

## PATENT ABSTRACTS OF JAPAN

(11)Publication number : 05-239326

(43)Date of publication of application : 17.09.1993

(51)Int.Cl.

C08L 67/02  
C08K 3/00  
C08K 3/22  
C08K 7/02  
C08L 25/18  
// (C08L 67/02  
C08L 27:18 )

(21)Application number : 04-043371

(71)Applicant : TEIJIN LTD

(22)Date of filing : 28.02.1992

(72)Inventor : SAEGUSA SHINJI  
OGAWA YOSHINARI  
SHIMOMA AKIRA

## (54) FLAME-RETARDANT RESIN COMPOSITION

## (57)Abstract:

PURPOSE: To provide the subject polyester resin composition excellent in mechanical properties, flame retardancy and fluidity (moldability) and low in flash.

CONSTITUTION: The objective flame-retardant resin composition is composed of 20 to 80wt.% polyethylene terephthalate (based on the whole composition, hereinafter used in the same mean), ≤60wt.% inorganic filler, 2 to 25wt.% brominated flame-retardant, 2 to 15wt.% antimony-based flame retardant and 0.05 to 10wt.% polytetrafluoroethylene resin produced by emulsion polymerization method.

## LEGAL STATUS

[Date of request for examination] 06.05.1997

[Date of sending the examiner's decision of rejection] 07.12.1999

[Kind of final disposal of application other than the  
examiner's decision of rejection or application  
converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of  
rejection]

BEST AVAILABLE COPY

[Date of requesting appeal against examiner's  
decision of rejection]

[Date of extinction of right]

Copyright (C); 1998,2003 Japan Patent Office

## \* NOTICES \*

JPO and NCIPi are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

---

CLAIMS

---

[Claim(s)]

[Claim 1] The flame-retardant-resin constituent which consists of 0.05 - 10 % of the weight (all inside of a constituent) of polytetrafluoroethylene resin manufactured by 20 - 80 % of the weight (all inside of a constituent) of polyethylene terephthalate, 60 % of the weight (all inside of a constituent) of inorganic bulking agents, 2 - 25 % of the weight (all inside of a constituent) of bromination polystyrene, 2 - 15 % of the weight (all inside of a constituent) of antimony system fire retardancy assistants, and the emulsion-polymerization method.

---

[Translation done.]

## \* NOTICES \*

JPO and NCIP are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

---

DETAILED DESCRIPTION

---

## [Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to a fire-resistant polyethylene-terephthalate-resin constituent. It excels in mechanical strength, fire retardancy, a fluidity, an appearance, etc. in more detail, and is related with a resin constituent with little weld flash.

[0002]

[Description of the Prior Art] Polyethylene terephthalate is conventionally used for extensive applications, such as an automobile, and electrical and electric equipment, electronic parts, as engineering plastics taking advantage of the outstanding thermal resistance, the mechanical characteristic, and the electrical property.

[0003] The constituent which mainly blended the organic halogen series flame retardant and the antimony system fire retardancy assistant to the application as which the fire retardancy of an electrical part etc. is generally required, and gave fire retardancy has been used. Moreover, many amount of macromolecules type bromine system flame retarders are used also in the flame retarder now for a mechanical strength, fire-resistant improvement, or the improvement in a surface appearance.

[0004] And the property required of resin in connection with the miniaturization of electronic parts, such as a switch, a connector, and a relay, and thinning is also becoming severe, and recently requires properties, such as the outstanding mechanical property and not only electrical characteristics but a high fluidity, low weld flash nature, etc., increasingly.

[0005] In order to give these functions to a resin constituent, selection of the additive appropriate for the purpose becomes important. Especially a flame retarder also has comparatively many additions also in an additive, and it is one of the additives which have large influence on the property of a resin constituent. As a bromine system flame retarder above-mentioned amount of giant molecules type, it divides roughly into a current aromatic polyester system resin constituent, and four kinds of flame retarders, such as a bromination bisphenol A mold polycarbonate resin system, a bromination bisphenol A epoxy resin system, a bromination polystyrene resin system, and a bromination polyphenylene ether resin system, are used for it. In order to give a high fluidity to a resin constituent among these, a bromination polystyrene resin system is best and lacks in a fluidity in other constituents. But although a fluidity improves, as for bromination polystyrene resin, there is no \*\*\*\*\* effectiveness in control of weld flash. Then, development of the fire-resistant polyethylene terephthalate system resin constituent with which can be satisfied of a fluidity and low weld flash nature has been desired.

[0006]

[Means for Solving the Problem] This invention was equipped with the outstanding mechanical property, fire retardancy, and a fluidity, and as a result of repeating examination in order to obtain a flame-retardant-resin constituent with little weld flash, it reached this invention.

[0007] 0.05 - 10 % of the weight (all inside of a constituent) of namely, polytetrafluoroethylene resin which manufactured this invention by (A) polyethylene terephthalate 20 - 60 or less (all inside of a constituent) % of the weight of 80-% of the weight (all inside of constituent), and (B) inorganic bulking agents, 2 - 25 % of the weight (all inside of a constituent) of (C) brominated flame retardants, 2 - 15 % of the weight (all inside of a constituent) of (D) antimony system fire retardancy assistants, and (E) emulsion-polymerization method -- since -- it is the becoming flame-retardant-resin constituent.

[0008] Hereafter, each constituent of the flame-retardant-resin constituent of this invention is explained in detail.

[0009] First, (A) polyethylene terephthalate (it may be written as "PET" below) used for this invention is an esterification reaction or a polymer which is made to carry out an ester exchange reaction, is subsequently made to carry out a polycondensation reaction, and is obtained about the dicarboxylic acid with which for example, at least 90-

mol % consists of a terephthalic acid or its ester plasticity derivative component, and the glycol component whose at least 90-mol % is ethylene glycol.

[0010] As an ester plasticity derivative of dicarboxylic acid, the dialkyl ester of a terephthalic acid, diaryl ester, etc. are mentioned preferably. As dicarboxylic acid which can be used together with a terephthalic acid in not more than 10 mol % as a dicarboxylic acid component, they are a phthalic acid, isophthalic acid, an adipic acid, a sebacic acid, and naphthalene, for example. - They are 1 and 4- or 2, 6-dicarboxylic acid, and diphenyl ether. - 4 and 4'-dicarboxylic acid etc. is mentioned.

[0011] Moreover, as a glycol which can be used together in not more than 10 mol % to ethylene glycol as a glycol component, glycols, such as propylene glycol, butylene-glycol, neopentyl glycol, cyclohexane dimethanol, 2, and 2-screw (4-hydroxyphenyl) propane, can be mentioned. In addition, as other components which can be used in case polyethylene terephthalate is obtained, oxy acid, such as p-hydroxybenzoic acid and a p-hydroxy ethoxy benzoic acid, can be mentioned.

[0012] Although especially the polyethylene terephthalate used by this invention is not limited about the polymerization degree, its intrinsic viscosity (inside of 35 degrees C and orthochromatic chlorophenol) is [ the thing of 0.4-1.0 ] desirable.

[0013] In the polyethylene-terephthalate-resin constituent of this invention, the above-mentioned polyethylene terephthalate occupies 20 - 80 % of the weight among [ all ] a constituent. The amount of polyethylene terephthalate becomes difficult [ fabrication nature ] at less than 20 % of the weight, and if it, on the other hand, exceeds 80 % of the weight, a problem will arise in the mechanical strength of mold goods.

[0014] Next, in this invention, it blends in order to obtain the mold goods which were excellent in engine performance, such as a mechanical strength, thermal resistance, and dimensional stability, in the (B) inorganic bulking agent.

[0015] As said inorganic bulking agent, tabular bulking agents, such as powder-like bulking agents, such as fibrous bulking agents, such as a glass fiber, a graphite fiber, an aramid fiber, a silica fiber, and an alumina fiber, and carbon black, a silica, a glass bead, a calcium silicate, a kaolin, wollastonite, and a mica, a glass flake, etc. can be used.

[0016] These inorganic bulking agents can use together one sort or two sorts or more.

[0017] It is desirable to use together a convergence agent or a finishing agent in use of these inorganic bulking agents, if required. If this example is shown, they will be functionality compounds, such as epoxy compound, an isocyanate system compound, a silane system compound, and a titanate compound.

[0018] These compounds may be beforehand used for an inorganic bulking agent by performing surface treatment or convergence processing, or may be added to coincidence in the case of ingredient preparation.

[0019] In this invention, the loadings of said inorganic bulking agent are 60 or less % of the weight, and are 5 - 50 % of the weight preferably. If the loadings of an inorganic bulking agent exceed 60 % of the weight, fabrication will become difficult, and also a problem arises in the mechanical strength of mold goods. Moreover, the amount of the functionality finishing agent used together is 0.05 - 5 % of the weight preferably one to 10% of the weight to an inorganic bulking agent.

[0020] Furthermore, it is blended in order that the bromination polystyrene of the (C) component may give fire-resistant effectiveness in this invention. The loadings of these brominated flame retardants are 2 - 25 % of the weight in [ all ] a constituent, and are 5 - 20 % of the weight preferably. Since a mechanical strength will fall if the fire-resistant effectiveness of mold goods that the loadings are obtained at less than 2 % of the weight is not acquired but it, on the other hand, exceeds 25 % of the weight, it is not desirable.

[0021] Moreover, the antimony system fire retardancy assistant of the (D) component used by this invention is blended in order to raise fire-resistant effectiveness further as a fire-resistant assistant of (C) brominated flame retardants. and some antimony pentoxide shown as this antimony system fire retardancy assistant by the antimony trioxide, antimony tetroxide, antimony pentoxide, and general formula  $(\text{NaO})_p\text{-Sb}_2\text{O}_5$  and  $\text{QH}_2\text{O}$  (p is 0.4-0.9, and Q is water of crystallization and shows the value of 0-4) -- sodium salt, sodium antimonate, etc. are mentioned preferably, and sodium antimonate is the most desirable especially.

[0022] The loadings of said antimony system fire retardancy assistant of this invention are 2.0 - 15 % of the weight in [ all ] a constituent, and are 3.0 - 10 % of the weight preferably. The loadings become insufficient [ less than 1 % of the weight / fire retardancy ], at 15 % of the weight or more, disassembly of resin or a compounding agent is promoted and mechanical strength falls.

[0023] (E) polytetrafluoroethylene resin used by this invention is manufactured by the emulsion-polymerization method, and it is used in order to reduce weld flash. The particle size of this resin has desirable 0.05-0.5 micrometers at a primary particle. Moreover, unless the weld flash reduction effectiveness is spoiled, it is also possible to carry out little copolymerization of the monomers, such as perfluoroalkyl vinyl ether. An addition is 0.05 - 10 % of the weight.

A fluidity will be spoiled, if there is no effectiveness of weld flash reduction and it exceeds 10 % of the weight, when there are few additions than 0.05 % of the weight.

[0024] Since a desired property is further given to the polyethylene-terephthalate-resin constituent of this invention according to the purpose, of course, it is also possible to blend a well-known additive, for example, lubricant other than the aforementioned component, a nucleating additive, a release agent, an antistatic agent, a surfactant, a plasticizer, a coloring agent, a heat-resistant stabilizer, UV stabilizer, an impact amelioration agent, a crystallization accelerator, etc.

[0025] The polyethylene-terephthalate-resin constituent of this invention can be easily prepared by the well-known facility and well-known approach which are generally used as the conventional resin constituent method of preparation. the approach of carrying out \*\*\*\* extrusion with an extruder, preparing a pellet, and fabricating the appropriate back, after mixing 1 each component and 2 -- the pellet with which presentations once differ is prepared and the approach of carrying out specified quantity mixing of the pellet, presenting shaping, and obtaining the mold goods of the purpose presentation after shaping, the approach of teaching 1 of each component or 2 or more directly to three making machines, etc. can use all. [ for example, ] Moreover, it is an approach desirable when performing homogeneity combination of these components to mix a part of resinous principle with components other than this as fine fine particles, and to add.

[0026] Hereafter, an example explains this invention still more concretely.

[0027] Moreover, the measuring method of characterization shown in the example is as follows.

[0028] \*\* Physical-properties measuring method tension test ASTM D It is conformity Izod impact strength to 638.

ASTM D It is conformity [0029] to 256. \*\* The stagnation trial in a making machine (thermal stability)

It fabricated, after making a sample pile up in a making machine cylinder for 30 minutes at 280 degrees C of cylinder temperatures, and the tensile strength of a shaping test piece was measured, and it considered as thermal stability and the standard which evaluates extent of degradation.

[0030] \*\* An inflammable test (UL-94)

According to the approach of the subject 94 (UL-94) of Underwriters Laboratories, it examined about fire retardancy using five test pieces (thickness: 1/32 inch).

[0031] \*\* It fabricated on the following conditions using the fluid trial metal mold for measuring method rod flow length measurement (mold cavity : 0.7mm in width-of-face [ of 10mm ] x thickness), and the fluidity was evaluated from the flow length (mold-goods die length).

process condition: -- cylinder-temperature: -- 280-degree-C injection pressure : 100kg/cm<sup>2</sup> die temperature : 80

degrees C [0032] \*\* The mold-goods visual-inspection disk (the diameter of 100mm, thickness of 3mm) was fabricated, the existence of a unique spot and extent were observed visually, and it judged according to the following ranks.

spot: -- O-:-less \*\*: -- a few -- it is -- those with x:large number [0033] \*\* It considered that the mold goods of bar flow length with a weld flash evaluation thickness of 0.05mm were weld flash, and asked for the weld flash characteristic as compared with bar flow length with a thickness of 0.5mm. A process condition is the cylinder temperature of 280 degrees C, the die temperature of 120 degrees C, and the 400 kgf/cm injection pressure 2. And it is injection-speed 100 mm/min (it is shown that there is so little weld flash that a characteristic is low).

Weld flash characteristic =(bar flow length with a thickness of 0.05mm)/(bar flow length with a thickness of 0.5mm) x100[0034]

[Examples 1-9 and the examples 1-9 of a comparison] After adding the various components shown in Table 1 or 2 to the polyethylene terephthalate of intrinsic viscosity (inside of 35 degrees C and orthochromatic chlorophenol) 0.7 and mixing to it, the constituent of a pellet type was prepared using the extruder. Subsequently, using this pellet, from this pellet, various test pieces were created with injection molding, and said evaluation was performed. The result was shown in Table 1 or 2.

[0035]

[Table 1]

組成	単位	実施例1	実施例2	実施例3	実施例4	実施例5	実施例6	実施例7	実施例8	実施例9
(A) ポリエチレンテレフタレート	重量%	56.0	46.0	56.0	56.0	56.0	56.0	56.95	56.7	52.0
(B) ガラス繊維 マイカ	"	30.0	20.0	30.0	30.0	30.0	30.0	30.0	30.0	30.0
(C) 臭素化ポリスチレン	"	—	20.0	—	—	—	—	—	—	—
(D) $Sb_2O_3$	"	10.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0
$Sb_2O_4$	"	3.0	—	—	—	—	—	—	—	—
$Sb_2O_5$	"	—	—	3.0	—	—	—	—	—	—
$(Na_2O)_{0.75}Sb_2O_5$	"	—	—	—	3.0	—	—	—	—	—
$NaSbO_3$	"	—	3.0	—	—	3.0	3.0	3.0	3.0	3.0
(E) ポリテトラフルオロエチレン (乳比重合) #	"	1.0	1.0	1.0	1.0	1.0	1.0	0.05	0.3	5.0
	"	—	—	—	—	—	—	—	—	—
品質	$kg/cm^2$	2300	1600	2400	2300	2100	2300	2200	2300	2400
引張強度	%	5.9	4.7	5.8	5.6	5.4	5.7	5.8	5.7	5.6
引張伸度	$kg \cdot cm/cm^2$	90	65	85	87	78	88	86	85	91
アイゾット衝撃強度 (ノッチ無)	$kg/cm^2$	2150	1500	2300	2200	2000	2300	2150	2200	2300
成形機内滞留試験 (引張強度)	—	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
熱安定試験 (UL94)	mm	440	300	410	400	470	430	410	450	400
流動性	—	○	○	○	○	○	○	○	○	○
成形品外観試験 (斑点)	—	1.2	1.4	1.2	1.3	1.3	1.3	2.0	1.7	1.0
バリ試験	—	—	—	—	—	—	—	—	—	—

[0036]

[Table 2]

組成	単位	比較例1	比較例2	比較例3	比較例4	比較例5	比較例6	比較例7	比較例8	比較例9
(A) ポリエチレンテレフタレート	重量%	64.5	39.0	57.0	56.0	57.5	42.0	56.97	45.0	52.0
(B) ガラス繊維 マイカ	"	30.0	30.0	30.0	30.0	30.0	30.0	30.0	30.0	30.0
(C) 臭素化ポリスチレン	"	—	—	—	—	—	—	—	—	—
(D) $Sb_2O_3$ $Sb_2O_4$ $Sb_2O_5$ $(Na_2O)_{0.175}Sb_2O_5$ $NaSbO_3$	"	1.5	27.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0
(E) ポリテトラフルオロエチレン (乳化重合)	"	3.0	3.0	3.0	3.0	1.5	17.0	3.0	3.0	3.0
" (懸濁重合)	"	1.0	1.0	—	—	1.0	1.0	0.03	12.0	—
品質	"	—	—	—	1.0	—	—	—	—	5.0
引張強度	kg/cm <sup>2</sup>	2500	1000	2300	1800	2200	1200	2400	2500	1200
引張伸度	%	5.1	4.7	5.5	4.3	5.0	4.5	5.2	5.0	4.5
アイゾット衝撃強度 (ノッチ無)	kg-cm/cm <sup>2</sup>	93	50	85	65	75	50	85	93	60
成形機内滞留試験 (引張強度)	kg/cm <sup>2</sup>	2400	600	2100	1600	2100	800	2200	2350	1000
燃焼性試験 (UL94)	—	V-2	V-0	V-0	V-0	V-2	V-0	V-0	V-0	V-0
流動性	mm	440	530	410	420	380	510	450	110	510
成形品外観試験 (斑点)	—	○	△	○	○	○	△	○	○	△
バリ指数	—	1.4	2.7	5.5	3.5	1.4	1.5	3.8	0.7	4.2

[0037]

[Effect of the Invention] The flame-retardant-resin constituent of this invention has the function in which the property shown below has been improved effectively.

1. It has the outstanding fire retardancy.
2. There is little heat deterioration of mold goods and there are also few falls on the strength.
3. Mechanical properties, such as tractive characteristics and Izod impact strength, are excellent.
4. There is very little weld flash of mold goods.



[Translation done.]

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☒ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☒ **FADED TEXT OR DRAWING**
- ☒ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☒ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**